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The potential for using geopolymers concrete in the UK

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Geopolymers are a novel class of inorganic polymers, which have the potential to replace Portland cement in a number of different applications. Geopolymers can utilise a higher level of industrial by-products than Portland cement blends and numerous studies have concluded geopolymer concretes have significantly lower embodied carbon dioxide than Portland-cement-based concretes. This paper examines the potential for the use of geopolymer binders as a Portland cement replacement in the UK. The quantities of material required, the major sources of these materials, the environmental implications and the barriers to implementation are discussed.

1. Introduction

Geopolymers are a novel class of inorganic binders and cements that are formed by activating an aluminosilicate powder precursor with an alkaline hydroxide or silicate solution (Davidovits, 2011). The precursors can come from a range of aluminosilicate sources with various ratios of aluminium (Al) and silicon (Si) and include natural minerals, calcined clays and industrial by-products (Davidovits, 2011; Duxson *et al.*, 2007). The term ‘geopolymer’ is very broad and can be used to describe a range of materials with a similar underlying chemistry, but this paper is focused on geopolymers in high-volume applications, where they can be used as either a replacement binder in concrete or as an alternative to other high-energy processes (e.g. as an alternative to firing bricks). This paper does not describe the underlying chemistry and manufacture of geopolymers in detail because this information is available elsewhere (e.g. Davidovits, 2011; Duxson *et al.*, 2007; van Deventer *et al.*, 2010).

Geopolymers are used as an alternative to Portland cement (PC)-based binders partly because of low embodied carbon dioxide compared with PC alternatives and, depending on economies of scale, for cost reasons (Davidovits, 1993; Gartner, 2004; McLellan *et al.*, 2011). However there can be additional technical merits to their use, including improved fire and acid resistance (Davidovits, 2011; Duxson *et al.*, 2007; Kong and Sanjayan, 2010).

2. Background

Although the term ‘geopolymer’ was used for alkali-activated clay-based binders in the 1970s (Davidovits, 2011), the first

refereed journal publication describing these materials was published in 1991 (Davidovits, 1991). There were earlier publications on alkali-activated binders, including the first publication on alkali-activated fly ash–slag binders, which originated from the UK (Purdon, 1940), but the earlier work was focused on high-calcium binders rather than the calcium-free geopolymers promoted by Davidovits (Duxson *et al.*, 2005). The current paper focuses mainly on materials with a low calcium content and uses the framework in Figure 1, which is based on the framework by RILEM Technical Committee 224-AAM (Provis, 2013).

Depending on the precursors and mix design, geopolymer concrete can appear similar to PC-based concrete in both fresh and hardened states, but the underlying chemistry and structure of the two are fundamentally different. Geopolymers are composed of an amorphous three-dimensional network comprising mainly aluminium (Al) and silicon (Si) oxides, whereas PC has a semi-crystalline gel binding phase (Davidovits, 2011; Lecomte *et al.*, 2006).

3. Current material use

To assess the potential for geopolymers in bulk construction applications, it is necessary to quantify the market for existing similar products. The data shown in Figure 2 were obtained from UK government statistics (BIS, 2012) and include UK material use for cement, concrete block and clay brick. The relative price increase from 2005 is indicated based on a value

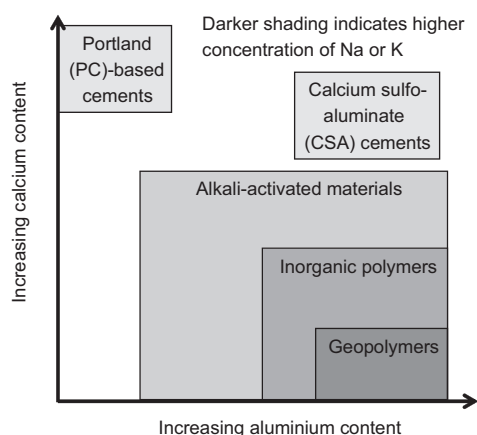


Figure 1. Classification of different subsets of alkali-activated materials with comparison to PC and calcium sulfo-aluminate cements (after van Deventer *et al.* (2010))

of 100 in 2005 for each of the materials. The material quantities were calculated based on representative densities of 1800, 1300 and 700 kg/m³ for the dense, lightweight and aerated concrete blocks and based on a fired brick unit mass of 2.35 kg, as the quantities are provided in inconsistent units in the original reference.

Some of the cement produced would have been used in the production of the concrete blocks as shown in Figure 2, but the majority would have been used for other applications, including bulk concreting and precast concrete component

production. Based on the data in Figure 2, the reduction in demand from the 2007 economic downturn did not create a corresponding reduction in cost, with prices generally increasing above the rate of inflation. The composite price index from the Bank of England indicates relative costs increasing from 100 to 119 between 2006 and 2011.

4. Potential precursor sources

Replacing significant quantities of PC with geopolymer binder with lower embodied carbon dioxide could have a significant impact on UK carbon dioxide emissions, but a number of technical and economic requirements must first be met, and there must be sufficient material to use as precursors.

The EU has developed a list of 14 minerals that are economically important but subject to a higher risk of interruption of supply, and the use of these minerals is inappropriate for bulk material applications. They are listed in alphabetical order in Table 1 (EC, 2010), with only magnesium having direct applicability to realistic PC alternatives currently under consideration. Details on other PC alternatives are beyond the scope of this paper and further information can be obtained elsewhere (e.g. Gartner, 2004; Juenger *et al.*, 2011). With critical minerals excluded from potential geopolymer precursors, alternatives could be either geologically based (natural) minerals, or industrial by-products.

4.1 Geologically based minerals

Much of the original work on geopolymers by Davidovits focused on metakaolin, which is relatively pure kaolin (also called china clay) that has been ‘calcined’ or heated until dehydroxylation. Kaolin is a 1:1 clay mineral with one layer of

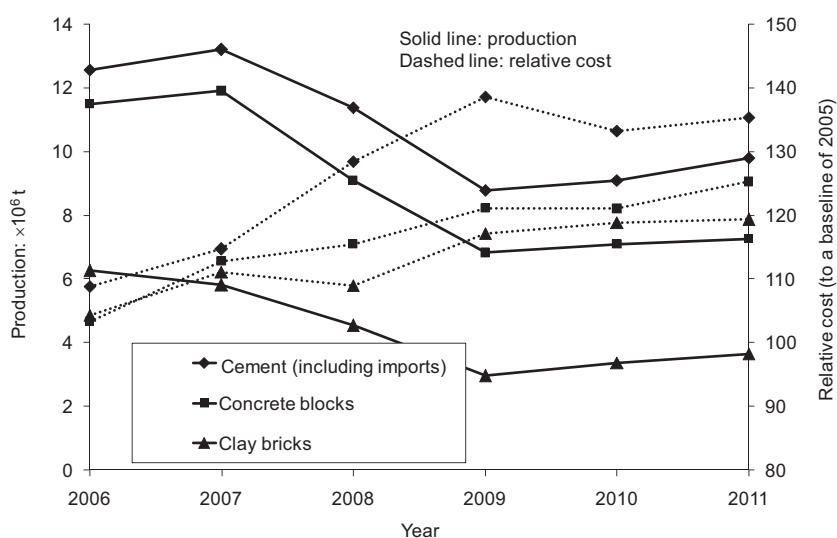


Figure 2. UK material production quantity and relative costs (data from BIS (2012))

Antimony	Indium
Beryllium	Magnesium ^a
Cobalt	Niobium
Fluorspar	PGMs (platinum group metals)
Gallium	Rare earths
Germanium	Tantalum
Graphite	Tungsten

^aMagnesium is regarded as critical, but magnesite (magnesium carbonate) was investigated and not included on the list of critical minerals.

Table 1. List of critical raw materials or mineral groups at EU level (EC, 2010)

silicate and another of alumina. For applications in geopolymers or as a pozzolan in PC-based mixes, the kaolin is normally heated to approximately 750°C and ground to form metakaolin. This heating and grinding does increase the embodied energy and carbon dioxide of the material, but as there is no carbon present in the kaolin, carbon dioxide is not directly emitted as with lime or cement manufacture. Geopolymers based on metakaolin can achieve the same strengths as commercially available PC concretes, but the fresh properties may differ. To achieve the rheology required for bulk concreting applications, high water contents are required for metakaolin-based geopolymers (e.g. Provis *et al.*, 2010) and this can have a detrimental impact on strength and durability. There is, however, scope to use metakaolin-based geopolymers in semi-dry applications, as discussed later.

The UK was the sixth largest global kaolin producer in 2009, with almost all extraction in the Cornwall/Devon area. Annual UK kaolin extraction reduced considerably between 2005 and 2009 (from 1.9 to 1.1×10^6 t) because of the high cost of UK extraction, whereas the fifth largest global producer in 2009, Iran, increased production from 0.5 to 1.2×10^6 t over the same period (BGS, 2011). The size of UK reserves is not published because of the commercial nature of this information, but it has been estimated that more than 50 years' capacity is available using current technology (BGS, 2011). UK china clay production peaked in 1988 at nearly 2.8×10^6 t, but only a small portion of this would have been processed to metakaolin, with the major uses as paper fillers and ceramics. Based on the difference between the peak and current capacity, it is estimated that an additional 1.6×10^6 t of dry kaolin could be sourced per year for geopolymer production, which would amount to approximately 1.4×10^6 t of metakaolin.

In addition to the relatively pure kaolin described above, the UK possesses considerable mixed and impure kaolin reserves

used for brick manufacture. UK brick production has been in decline since the 1970s, with almost 16×10^6 t of brick clay extracted in 1974 (BGS, 2007), decreasing to 4×10^6 t in 2011, as shown in Figure 2. What the brick industry describes as brick clay is not geologically clay, but mainly unreactive silt and sand with approximately 30% potentially reactive clay-sized particles (Heath *et al.*, 2009). The clay minerals preferred in brick manufacture are kaolinite and illite (BGS, 2007) and both have been successfully used for geopolymer production. Taking the 12×10^6 t difference in annual extraction between 1974 and 2011 and estimating the clay content of this as 30% of the total mass, there should be at least 3.6×10^6 t of mainly kaolin and illite available per annum from current and inactive brick extraction pits.

In addition to the kaolin extracted as almost pure 'china clay' and that used in brick manufacture, there are a number of other sources of kaolin and other suitable clays, including from 'ball clay' extracted in the Devon area, and extraction from other weathered materials and quarry or mining wastes throughout the UK. At the time of writing, one UK company is actively marketing a range of geopolymer building products and these include weathered basalt precursors (containing kaolin).

4.2 Industrial by-products

Although geologic minerals suitable for use in geopolymers are available in very large quantities, recent research into geopolymers has focused on the use of industrial by-products, notably fly ash and slags (Davidovits, 2011; Duxson *et al.*, 2005, 2007; Gartner, 2004). Geopolymer concretes based on fly ash and slags can have similar workability to concretes based on PC, and these geopolymers may not require heating during curing (Davidovits, 2011). Fly ash and slags have the benefit of high temperatures during manufacture, which can promote an amorphous form that is utilised when used as an additive to PC blends and when forming geopolymers. This removes the need for dehydroxylation, which is required for geologically based geopolymer precursors.

Approximately 6×10^6 t of fly ash or pulverised fuel ash (PFA) is produced in the UK each year, of which approximately 50% is sent to landfill (McCarthy *et al.*, 2008; WRAP, 2008). In addition, there are significant stockpiles of PFA of variable quality around the UK, with approximately 114×10^6 t accessible in stockpiles. PFA has been extensively used as a precursor in geopolymer production and this is probably the most significant current UK resource for geopolymer production. Although no comprehensive research programme has been conducted on the suitability of current UK PFA and PFA reserves for geopolymer production, research in 2008 indicated 55% of reserves would be suitable as a binder replacement in PC blends, but processing may be required to separate the resource into fractions for various uses (McCarthy *et al.*, 2008). Some sources of PFA that are suitable for PC blends are not

ideal for geopolymers, and vice versa, but for the purposes of this analysis it is assumed that a similar percentage is available for geopolymer precursors.

The use of PFA as a binder replacement in PC-based concrete is governed by standard EN 450: Fly ash for concrete (BSI, 2012), and this includes specifications for PFA from co-combustion of coal and biomass, with a minimum 60% coal by dry weight in the fuel mix, or 50% if the biomass is green wood. This presents a particular challenge in the UK context as government energy policy indicates a significant decrease in coal-based electricity supply and a corresponding increase in renewable supply, as shown in Figure 3 (based on DECC (2011)). The figure combines conventional and limited carbon capture and storage (CCS) for both coal- and gas-based electricity, and includes electricity from combined heat and power (CHP) plants. The renewable category includes all forms of renewable energy, including biomass, solar and wind. It is predicted that biomass will account for 25–45% of the renewable generation by 2020. The use of biomass ash in geopolymers shows particular promise because biomass ash is generally higher in potassium than coal ash (Steenari *et al.*, 1999), potentially reducing the activator required. The activator provides the greatest single contribution to the embodied carbon dioxide of geopolymer concretes (Habert *et al.*, 2011; McLellan *et al.*, 2011) and minimising activator use can therefore have environmental benefits.

A variety of ferrous and non-ferrous slags have been used in geopolymers (Komnitsas *et al.*, 2011). The main slags currently

produced in the UK are blast furnace and steel slag, although the non-ferrous metal industry in the UK has produced aluminium, copper, lead, magnesium, nickel, titanium and zinc slags (Capita Symonds, 2007). Of the non-ferrous metals, only lead and aluminium slags are still produced in significant quantities from primary processing. Although primary zinc processing has ceased in the UK, there is reportedly a 2×10^6 t stockpile of zinc slag near Avonmouth (Capita Symonds, 2007). Although this material may be suitable for geopolymerisation, there may be insufficient material to justify the capital cost of a processing plant. The use of non-ferrous and steel slags as geopolymer precursors is an area requiring further investigation and these are not therefore included in subsequent analysis.

The majority of research into slag-based geopolymers is focused on ground granulated blast-furnace slag (GGBS) (Davidovits, 2011; Duxson *et al.*, 2007; Gartner, 2004). As the calcium content in GGBS is higher than for other common geopolymer precursors, it may fall outside the 'geopolymer' block in Figure 1, but as a very promising alkali-activated material it is included. Approximately 2×10^6 t of GGBS are produced each year in the UK (Capita Symonds, 2007), and almost all of this is secured through long-term contracts between GGBS producers and PC manufacturers. This aspect is discussed in more detail later.

In addition to PFA and slags, there is potential for other smaller waste and by-product streams to be used, including paper sludge ash, which is particularly rich in amorphous aluminosilicates from the combustion of wood fibres and the

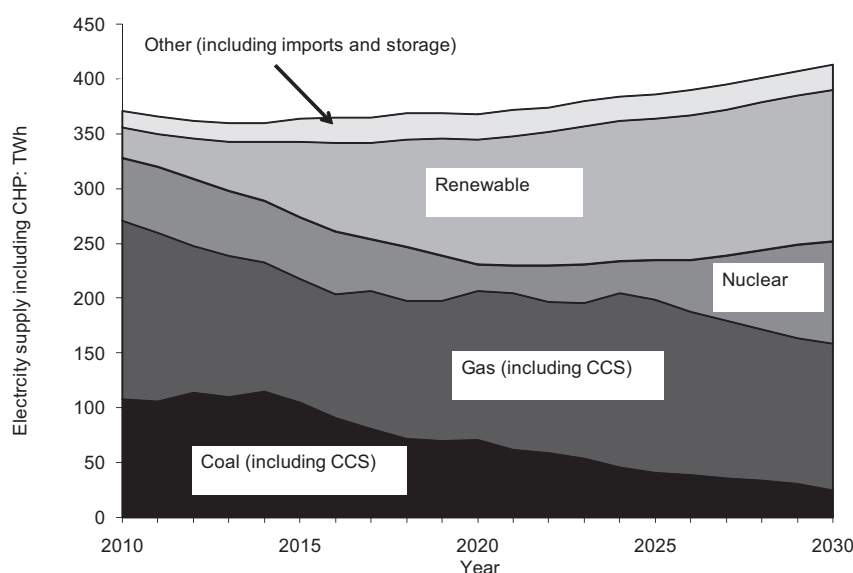


Figure 3. Predicted UK energy supply sources (based on DECC (2011))

dehydroxylation and calcination of kaolin and limestone fillers.

4.3 Total available materials

The current sources of major precursors are summarised in Table 2; these data are based on current production, noting that production of coal-based fly ash is predicted to decrease whereas biomass fly ash is expected to increase (Figure 3).

If Figure 2 and Table 2 are viewed together, it can be concluded that there are potentially sufficient precursors to replace all 10×10^6 t/year of current UK cement use with metakaolin-, PFA- or GGBS-based geopolymer cement if some existing fly ash stockpiles are used. Because of the different workability of clay-based geopolymers (Provis *et al.*, 2010), it is likely that PFA and/or slag-based geopolymers would be more suited for in-situ bulk concrete applications, while geologically based geopolymers could be utilised for semi-dry pressed blocks and other precast elements where alternative compaction methods (including extrusion) are available and where it is easier to control curing.

There are, however, a number of reasons why a complete replacement of PC mixes is unlikely, and these include the versatility and ease of use of PC compared with current formulations of geopolymer cements.

5. Environmental considerations

Because of space restrictions, this section is focused on use of waste materials and embodied carbon dioxide, because these are the focus of most of the existing literature on the environmental impact of geopolymers and other potential PC concrete alternatives (Davidovits, 1993; Gartner, 2004; Juenger *et al.*, 2011; McLellan *et al.*, 2011). Other environmental impacts should, however, also be considered when using geopolymers (Habert *et al.*, 2011).

5.1 Use of waste materials

The 2008 EU directive 2008/98/EC states that a material may be classified as a by-product rather than a waste if

- (a) further use of the substance or object is certain; (b) the substance or object can be used directly without any further processing other than normal industrial practice; (c) the substance or object is produced as an integral part of a production process; and (d) further use is lawful.

As mentioned earlier, almost all of the GGBS and half of the PFA produced in the UK is currently used. If PFA use remains constant and coal-based electricity supply follows the UK government predictions in Figure 3, it is likely that all new suitable PFA produced in the UK will be used within 10 years, and additional PFA will have to be extracted from stockpiles as in other European countries. Under these conditions, neither GGBS nor PFA should be considered waste materials.

Any ash from biomass co-fired with coal at ratios unacceptable to EN 450 will be considered a waste and will have to be disposed of, or a beneficial use for it found. There is potential for geopolymers to be based on PFA which does not meet the requirements of EN 450 (because of too high a biomass content or for other reasons) or on ashes from combustion of other renewable materials, including paper sludge ash, but this will require further investigation to confirm the suitability of these materials.

5.2 Embodied carbon

Concrete production accounts for approximately 7% of global carbon dioxide emissions (Basheer, 2009). The change from the almost universal use of PC-based concrete to a concrete with significantly lower embodied carbon dioxide would therefore have a marked effect on UK emissions. The extent of this reduction would depend on the availability, uptake and embodied carbon dioxide of the replacement concrete. All

Potential precursor	Quantity available: $\times 10^6$ t/year ^a	Potential geopolymer products
China clay/metakaolin ^b	At least 1.6	Cement for precast elements/blocks
Brick clay	At least 3.6	Geopolymer blocks
GGBS ^c	2	Cement for concrete/blocks
PFA ^d	3.3 + 57 in stockpiles	Cement for concrete/blocks

^aReferences included in text.

^bBased on difference between peak and current production.

^cAlmost all current production of 2×10^6 t/year used in PC based mixes.

^dCoal-based PFA is predicted to decrease significantly over the next 10–20 years, but biomass ash is likely to increase significantly.

Table 2. Summary of major potential precursors for geopolymers in the UK, based on current production

information presented here is based on simplified cradle-to-factory-gate analyses, because there is insufficient information on the full life cycle, including end of use, of geopolymer concrete or other geopolymer building products.

Rather than performing a full analysis of the embodied carbon dioxide of geopolymers, the percentage reduction in embodied carbon for the binder only or concrete from recent sources in the literature is summarised in Table 3. These references do not specifically include heating during curing because many modern geopolymer mixes do not require heat curing, which is often impractical for on-site applications.

As shown, all references show geopolymer concretes have lower embodied carbon dioxide than PC-based mixes. Because the embodied carbon dioxide data for sodium silicate used in all references were developed in 1995 for the European situation (Fawer *et al.*, 1999), average carbon dioxide emissions per unit electrical and heat energy have reduced by over 20% (IEA, 2011), which implies the benefit of using geopolymers may be even greater than indicated.

The reference with the lowest reduction (Habert *et al.*, 2011) assumed a neutral sodium silicate ($\text{SiO}_2\text{Na}_2\text{O}$ weight ratio of 3.3) activator instead of an alkali sodium silicate activator ($\text{SiO}_2\text{Na}_2\text{O}$ ratio of 2), which is more beneficial for geopolymerisation. This assumption artificially increases sodium silicate embodied carbon dioxide emissions by 48% (Fawer *et al.*, 1999). Because the sodium silicate contributed approximately 70% of the embodied carbon and with an error of almost 50% in this value, the calculated emissions in the Habert *et al.* (2011) paper are considered erroneously high and are not used in the subsequent analysis.

Based on the data in Table 3, and with PC-based concrete production accounting for approximately 7% of global carbon dioxide emissions, carbon dioxide reductions can be calculated based on the percentage PC concrete replacement with

geopolymer concrete as presented in Figure 4. The range is calculated using the maximum and minimum values from Table 3 (excluding Habert *et al.* (2011)), all of which include no allocation of emissions for PFA precursors. Although the allocation of emissions is a contentious issue for the concrete industry, this is the correct assumption for this particular analysis because the intention is to show the potential for change in carbon dioxide emissions and allocation of emissions has no direct effect on actual emissions. Allocation of emissions for GGBS and PFA is related to whether they are considered wastes or by-products and recent research by van den Heede and de Belie (2012) indicated an allocation of embodied carbon dioxide to PFA on a mass basis would result in unblended PC having lower emissions than a PFA/PC blend. The allocation of emissions is an area requiring further investigation in the context of geopolymers.

Although it is unlikely that there will be a 50% replacement of PC-based concrete with geopolymer concrete in the near future, the effect shown in Figure 4 would be considerable, with carbon dioxide savings approaching the carbon dioxide emissions of global aviation of approximately 3% (Mayor and Tol, 2010). The embodied carbon of geopolymers could be further decreased if lower embodied carbon sources of activators are found, which could include use of by-products in activators and lower-carbon carbon dioxide of power, the main component of the embodied carbon dioxide in sodium silicate activators (Fawer *et al.*, 1999). Because carbon dioxide is directly emitted during the manufacture of PC (approximately 0.55 t carbon dioxide chemically emitted per tonne PC produced, with the remainder being processing emissions), there is little scope for these large reductions in embodied carbon dioxide for PC.

6. Barriers to geopolymer use

UK coal-based PFA production is expected to decrease, so there may be insufficient PFA to use as precursors in the future unless the increasing biomass ash resource is utilised. If the anticipated decrease in PFA is realised, a similar situation to

Reference	Location	Percentage embodied carbon compared with PC binder or concrete
McLellan <i>et al.</i> , 2011	Australia	44–64
	France	55–82 (higher figure for economic allocation of emissions from fly ash)
Habert <i>et al.</i> , 2011 ^a		
Davidovits, 2011	France	19 (no emissions allocation for GGBS)
van Deventer <i>et al.</i> , 2010	Australia	20–40 (lower figure is for binder only)

^aOnly fly ash geopolymers included, although GGBS and metakaolin also investigated.

Table 3. Geopolymer emissions as a percentage of PC-based concretes or pastes

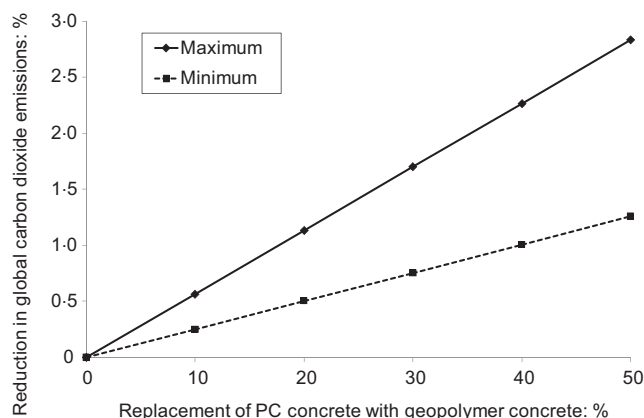


Figure 4. Effect of geopolymer concrete uptake on global carbon dioxide emissions

that with GGBS could develop, where all suitable new PFA is secured by PC manufacturers through long-term contracts. Because of the extensive stockpiles of PFA around the UK, it is unlikely this will happen for a number of years, but it could limit the potential use of innovative PC replacements containing GGBS or PFA. The effect of a potential supply chain monopoly by large PC manufacturers is an area requiring further investigation at a policy level, because it could have a detrimental effect on UK carbon dioxide emissions by stifling competition from a number of low-carbon-dioxide cements based on these materials.

At the time of writing, there is only one company in the UK extensively marketing geopolymer building products, but the market is more developed elsewhere, particularly in Australia. The UK construction industry is notoriously conservative, which is a barrier to innovation uptake. The Egan (1998) report on the UK construction industry stated: 'There is already frustration amongst component suppliers that their innovations are blocked because construction workers cannot cope with the new technologies that they are making available'. If geopolymers are formulated to be used in the same manner as conventional concretes, either through mix design or use of precast elements including blocks, acceptance by construction workers will be improved as they may not realise the difference. As mentioned earlier, the issue of high water contents for metakaolin-based geopolymers to achieve rheological properties similar to PC-based concrete (Provis *et al.*, 2010) may limit their use to precast and block applications. PFA- and GGBS-based geopolymers are therefore more likely to be suitable for bulk concreting applications. Demonstration projects can assist in gaining acceptance by construction workers.

Even if geopolymer concrete mixes and products are fully accepted by construction workers, designers are reluctant to

specify materials where no standards exist and where there are limited long-term performance data. This is a point of concern for geopolymer concretes where no nationally adopted standards currently exist, although RILEM committee 224 recently completed work on standardising performance testing procedures for these materials (Provis, 2013). Performance standards are not always compatible with EU approaches, where standard EN 206-1:2000 (BSI, 2000) for the 'Specification, performance, production and conformity of concrete' states that concrete must gain strength by hydration of cement on mixing with water. This limits the use of alternative cements that do not use hydration reactions, even if they demonstrate considerable environmental and/or technical benefit. The UK government has recently investigated these issues in the construction industry and has stated (Cabinet Office, 2011)

the criteria for value need to be converted into standards and specifications that can be passed to suppliers as part of the brief that they are required to meet. There should be consistency across Government in how these standards are set, and wherever possible they should be performance (or outcome) based, rather than prescriptive, so that opportunities for innovation by the supply side are maximised.

Although this indicates that the UK government has recognised the need for performance specifications, there are no indications that EU standards will follow this approach in the near future.

7. Conclusion

Geopolymers have the potential to decrease global carbon dioxide emissions significantly. Reductions of this magnitude are not available to PC manufacture because of the chemical release of carbon dioxide through the heating of limestone during production. Geopolymers can be based on abundant natural (mainly metakaolin) and industrial waste materials (mainly GGBS and PFA). Because of the ease of use, it is unlikely that PC will cease to be used in the UK, but there is potential for geopolymer binders to replace a significant portion of current PC used.

Initial routes to market for geopolymer concrete products in the UK could be by way of precast elements (including blocks), because the risks are lower and control of production and curing higher than for in-situ mixes. Education on geopolymers and other low-carbon concrete alternatives should be included across the full spectrum of the construction industry to aid uptake.

There may not be sufficient PFA meeting EN 450 and GGBS produced in the UK if geopolymer cement were to replace a significant portion of the current PC production, because coal

power generation is expected to decrease significantly. It is likely that PFA will have to be extracted from existing stockpiles to meet blended PC requirements within the next 10 years, and even earlier if geopolymer concretes based on PFA are extensively used. Almost all GGBS currently produced in the UK is already used, the majority in PC blends. The options are therefore either to continue to use PC-based concretes with a relatively low PFA/GGBS content (and with high carbon dioxide emissions), to import geopolymer precursors into the UK for geopolymer production, or to investigate other suitable geopolymer precursors.

Other potential precursors could include new or existing waste streams with suitable amorphous aluminosilicates, or geological alternatives. Potential new waste streams include non-ferrous slags, co-fired or biomass fly ash and paper sludge ash, but the use of these new waste streams in geopolymers requires extensive research. Geological alternatives should be investigated for the UK to ensure long-term security of supply, but these may only be suitable for limited applications.

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